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Constraining Holocene hydrological changes in the Carpathian-Balkan region using speleothem $\delta^{18}O$ and pollen-based temperature reconstructions

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Abstract

Here we present a new speleothem isotope record (POM2) from Ascunsă Cave (Romania) that provides new data on past climate changes in the Carpathian-Balkan region from 8.2 ka until present. This paper describes an approach towards constraining the effect of temperature changes on calcite $\delta^{18}O$ values in stalagmite POM2 over the course of the Middle Holocene (6–4 ka), and across the 8.2 and 3.2 ka rapid climate change events. Independent pollen temperature reconstructions are used to constrain the temperature-dependent component of total isotopic change in speleothem calcite. This includes the temperature-dependent composition of rain water attained during vapour condensation and during calcite precipitation at the given cave temperature. The only prior assumptions are that pollen-derived average annual temperature reflects average cave temperature, and that pollen-derived coldest and warmest month temperatures reflect the range of condensation temperatures of rain at the cave site. This approach constrains a range of values between which speleothem isotopic changes should be found if controlled only by surface temperature variations at the cave site. Deviations of measured $\delta^{18}O_c$ values from the calculated range are interpreted towards large-scale hydrologic change independent of local temperature.

Following this approach, we show that an additional 0.6 ‰ enrichment of $\delta^{18}O_c$ in the POM2 stalagmite was caused by changing hydrological patterns in SW Romania during the Middle Holocene. Further, by extending the calculations to other speleothem records from around the entire Mediterranean Basin, it appears that all Eastern Mediterranean speleothems recorded a similar isotopic enrichment due to changing hydrology, whereas all changes recorded in speleothems from the Western Mediterranean are fully explained by temperature variation alone. This highlights a different hydrological evolution between the two sides of the Mediterranean.

Our results also demonstrate that during the 8.2 ka event, POM2 stable isotope data fit the temperature-constrained isotopic variability, with only little hydrologic change at most. In the case of the 3.2 ka event, the hydrological factor is more evident. This

implies a potentially different rainfall pattern in the Southern Carpathian region during this event at the end of the Bronze Age.

This study brings new evidence for disturbances in Eastern Mediterranean hydrology during the Holocene, bearing importance for the understanding of climate pressure on agricultural activities in this area.

1 Introduction

The impact of Holocene rapid climate changes on human communities in the Eastern Mediterranean region was documented by Weninger et al. (2009). Staubwasser and Weiss (2006) showed that rapid climate change events can alter the hydrological cycle, putting pressure on agricultural societies and sometimes leading to their demise. This paper attempts to bring new information about the response of hydrology to temperature change in different parts of Europe, focusing on the Carpathian-Balkan region.

In the region surrounding the Eastern Mediterranean, proxy records suggest that conditions were more humid during the Early Holocene compared to present-day moisture budgets (Rossignol-Strick, 1999; Rohling et al., 2002). Enhanced freshwater flux roughly between 10 000 yr before present (10 ka) and 6 ka led to stratification and sapropel formation in the Eastern Mediterranean, whereas depleted $\delta^{18}O$ values in lacustrine calcareous microfossils and endogenic carbonate deposits suggest less evaporation across the region during that time compared to the present day. A stacked oxygen isotope record generated from lake proxies around the Eastern Mediterranean shows a general drying trend between 6 and 4 ka (Roberts et al., 2008). A pattern of increasing $\delta^{18}O$ values is documented in speleothem records from south-central Europe and the Eastern Mediterranean (McDermott et al., 2011, and references therein). This has been suggested by McDermott et al. (2011) to reflect less rainfall from Atlantic-sourced moisture reaching the Eastern Mediterranean region during the Late Holocene.

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Due to the topographic complexity and rather sparse data distribution, reports from across the Carpathian-Balkan region do not provide yet a unified view on past environmental change, but rather point to possibly contrasting Holocene hydroclimatic evolution at regional scale (Feurdean et al., 2008; Magyari et al., 2013). Lake records from the southern Balkans, such as Ioannina, Greece (Frogley et al., 2001) and Prespa and Ohrid, Macedonia (Leng et al., 2010) indicate high humidity throughout the Early Holocene, whereas paleolimnological records from Steregoiu, NW Romania (Feurdean et al., 2007) and Sfânta Ana Lake, central Romania (Magyari et al., 2009), suggest that lower humidity persisted in the area. At Sfânta Ana, a volcanic crater lake with no outflow, water levels began to rise only after 7.4 ka (Magyari et al., 2009).

Several Holocene speleothem records are available from the Romanian Carpathians (Onac et al., 2002; Tămaş et al., 2005; Constantin et al., 2007). Trends towards higher values seen in these time series throughout the Holocene were interpreted as reflecting rising temperatures. McDermott et al. (2011) followed on the interpretation of European speleothem records documenting decreasing rainout gradients across the Holocene on a longitudinal transect. However, a more specific distinction between hydrology and temperature-driven changes requires further research because interpreting stable oxygen isotope records from speleothems in terms of palaeoclimate is not generally straightforward (McDermott, 2004; Lachniet, 2009; Tremaine et al., 2011). For example, the effects of temperature and hydrologic changes may cancel each other out as cave temperature and rainfall temperature affect speleothem δ^{18}O values in opposite directions. Changes in seasonality of both rainfall and calcite precipitation are difficult to detect (Baker et al., 2011). Furthermore, moisture sources and transport trajectories, which generally affect the stable isotopic composition of meteoric water in Europe (Rozanski et al., 1982), may respond to regional-scale climate changes in contrast to local ones. Consequently, specific temperature or hydrological information is rarely directly quantifiable from speleothem stable isotope records. An example is the muted or even absent signal around the 8.2 ka cold event in speleothem δ^{18}O records throughout Romania (Tămaş et al., 2005; Constantin et al., 2007), despite this event

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being clearly identifiable in peat bog pollen records (Feurdean et al., 2007), in Balkan lake records (Pross et al., 2009; Panagiotopoulos et al., 2013), as well as in the Aegean Sea (Marino et al., 2009). This ambiguity of speleothem δ^{18}O records with respect to climatic events and transitions raises the question of how more specific information on the nature of climate change can be extracted from this proxy.

In this study we present a new speleothem isotopic record from Ascunsă Cave located on the eastern slopes of the Carpathian Mountains in Southern Romania (Fig. 1), in an area under periodical Mediterranean hydroclimate influences (Bojariu and Paliu, 2001; Apostol, 2008). We combine information from regionally averaged pollen-based temperature reconstructions from Europe across the Holocene (Davis et al., 2003) with the new oxygen isotope data from Ascunsă Cave, alongside a detailed comparison with published speleothem records from Romania (Onac et al., 2002; Tămaş et al., 2005; Constantin et al., 2007) and the Mediterranean (McDermott et al., 1999; Bar-Matthews et al., 2003; Drysdale et al., 2006; Vollweiler et al., 2006; Verheyden et al., 2008; Fleitmann et al., 2009). We also attempt to constrain the regional-scale hydrologic information inherent by speleothem δ^{18}O change across the Holocene, focussing on Mediterranean climate trends observable after 6 ka (Mayewski et al., 2004; Roberts et al., 2008, 2011; McDermott et al., 2011).

Finally, local pollen data sets (Feurdean et al., 2008; Bordon et al., 2009) are used to constrain selected rapid climate shifts associated with the 8.2 ka and the 3.2 ka events.

2 Materials and methods

2.1 Cave setting and stalagmite characteristics

Ascunsă Cave is located on the eastern slopes of Mehedinţi Mountains, Southern Carpathians (45.0° N, 22.6° E, 1050 m alt.) in south-western Romania (Fig. 1). It is a 400 m long and over 200 m deep contact cave developed by river erosion of

Turonian-Senonian wildflysch (mélange) below an Upper Jurassic-Aptian limestone cover (Codarcea et al., 1964).

The cave is well decorated with speleothems and throughout its course there is a chaotic mixture of collapsed blocks and speleothem fragments reflecting the undermining of the wildflysch walls by fluvial erosion or their failure to support massive flowstone formations.

The analysed stalagmite (POM2) is 77.4 cm long and composed of well-laminated and densely compacted white calcite (Figs. 3 and 4). Topographic survey at the cave site revealed that limestone thickness above the stalagmite sampling site is ~ 100 m.

2.2 Present day climatology of the study area and cave monitoring

The regional climate of the Romanian Carpathians is temperate-continental, characterised by a predominantly Atlantic origin of air masses (Baltă and Geicu, 2008). It is also influenced (in the south-western part) by Mediterranean cyclonic activity that is responsible for milder temperatures and increased winter rainfall in the area of the study site compared to northern or eastern Carpathians (Bojariu and Paliu, 2001; Apostol, 2008). Most of the cyclones affecting the study area originate in the central Mediterranean (around the Gulf of Genoa), but cyclones from the Aegean Sea also reach this region periodically (Apostol, 2008). Seasonal variation is observed in the formation of these cyclones, the southern shift of the polar jet stream in winter being linked to a stronger Mediterranean cyclogenesis during this season (Trigo et al., 2002).

Figure 2 illustrates the seasonal differences in precipitation recorded between 1961 and 2000 at two meteorological stations relevant for this study, Drobeta (SW Romania) and Stâna de Vale (W Romania) (data from Dragotă and Baciu, 2008). There is a clear difference in rainfall seasonality between the two regions, with Stâna de Vale having one rainfall peak in the summer, whereas at Drobeta two main rainfall periods are peaking in spring and early winter (Fig. 2).

Ascunsă Cave was monitored between July 2012 and November 2013 for atmospheric physical parameters and drip water isotopic composition. Temperature (T),

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relative humidity (RH) and CO_2 partial pressure (pCO_2) were measured at three sampling locations (POM A, POM2, and POM B) within Ascunsă Cave, using two Vaisala probes, GMP70 for pCO_2 and HMP75 for T and RH. Drip water collected from stalactite tips at the sampling sites was analysed for δ^{18}O and δD on a Picarro L2130-i Cavity Ring-Down Spectroscope at Babeą-Bolyai University (Cluj-Napoca, Romania) following the method described by Brand et al. (2009). The analytical precision is better than ±0.03 ‰ for δ^{18}O and ±0.07 ‰ for δD. For data normalization, two laboratory reference waters (VEEN and HTAMP) that were calibrated directly against VSMOW were measured repeatedly in each run. Results are expressed in ‰ on the VSMOW scale.

2.3 U-series dating and stable isotope analysis of speleothem samples

For U-Th dating, calcite samples were analysed on a THERMO Neptune MC-ICPMS following procedures outlined in Hoffmann et al. (2007) and Hoffmann (2008). In total, 14 U-Th samples were measured, covering the entire length of the stalagmite. Three pairs of samples were drilled immediately underneath and above visible changes in growth axis at 43.4, 54.4 and 63.9 cm.

A total of 150 stable isotope samples were hand drilled at 5 mm resolution using a 0.5 mm drill bit. All samples were analysed at the University of Oxford on a Thermo Delta V Advantage mass spectrometer equipped with a Kiel IV Carbonate Device. Results are reported relative to the Vienna Pee Dee Belemnite (VPDB) standard, and external precision on replicate samples (NBS 18, NBS 19, and a local carbonate standard) run daily on this system was 0.06 % for δ^{18}O and 0.03 % for δ^{13}C.

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3 Results and discussion

3.1 U-series dating results and growth model

The U-Th ages suggest that the stalagmite started growing around 17.2 ka, but most of the growth occurred between 8.2 ka and the present. The age model for the Holocene part of the stalagmite (Fig. 5) is based on eleven U-Th ages with typical dating uncertainties ranging between 1 and 6 % (2σ) (Table 1). The stalagmite was active at the time of sampling, thus the age at the top (77.4 cm) is assumed to be 0 (relative to 2009, the year of sampling) and is used as an additional tie point in the growth model calculation. The ^{238}U concentration varies between 18 and 50 ng g^{-1}, and ^{232}Th concentration ranges between 0.1 and 12.2 ng g^{-1}. Dating uncertainties are therefore mainly resulting from small U concentration and a significant correction for initial Th, combined with the young age of the stalagmite which yields low ^{230}Th/^{232}Th activity ratios (< 10) for six of the age determinations. Two samples (POM 09-2/III and POM 09-2/VI) are entirely dominated by detrital Th, with ^{230}Th/^{232}Th activity ratios < 0.5 and have not yielded resolvable U-Th ages.

We measured U and Th isotopes on a sample from the top of the actively forming stalagmite in order to assess a reliable correction factor. The results show a ^{238}U concentration of 18.3 ± 0.1 ng g^{-1} and a ^{232}Th concentration of 12.2 ± 0.1 ng g^{-1}. The measured ^{230}Th in the top sample is assumed to be entirely of detrital origin and the apparent age of 8.3 ka a result of initial thorium contamination. The ^{230}Th/^{232}Th activity ratio of this sample is 0.6 ± 0.05, which indicates detrital activity ratios for ^{230}Th/^{232}Th, ^{234}U/^{232}Th and ^{238}U/^{232}Th of 0.6 ± 0.05, if we assume the detritus to be in secular equilibrium. We note that this factor is well within the range of the bulk earth value of 0.8 ± 0.4 (Wedepohl, 1995). We therefore use the value of 0.6 with a conservative uncertainty of 50 % to correct for initial Th.

The growth model of stalagmite POM2 (Fig. 5) was generated using the StalAge algorithm of Scholz and Hoffmann (2011).

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3.2 Cave monitoring results

Monitoring data show a stable average temperature of 8.2 ± 0.6 °C at the stalagmite site. Relative humidity is also stable around 94 ± 2.5 % during the year, especially at sampling sites POM2 (where stalagmite POM2 was sampled) and POM B situated deeper inside the cave (Table 2).

Isotope measurements of drip waters at POM2 site show rather consistent values for both $\delta^{18}O$ (-10.57 ± 0.04 ‰) and δD (-70.58 ± 0.20 ‰), during the autumn–winter months. This may indicate an efficient mixing of waters in the aquifer, without capturing any individual rain events.

Analysis of calcite farmed on glass plates also revealed relatively constant values with mean $\delta^{13}C$ of -10.30 ± 0.8 ‰ and $\delta^{18}O$ of -7.91 ± 0.2 ‰ for both POM2 and an adjacent stalagmite, POM X (Table 3).

We constructed a local drip water line for Ascunsă Cave (Fig. 6) using $\delta^{18}O$ and δD values of drip waters from all three sampling sites. Compared to the global (GMWL) and Mediterranean (MMWL) meteoric water lines, the Ascunsă groundwater line (AGWL) is defined as $\delta D = 6.9 \times \delta^{18}O + 2$ and plots above the GMWL. This could indicate either the existence of enrichment processes at local scale or a mixture between humid Atlantic and drier Mediterranean vapour sources.

To test the existence of equilibrium fractionation conditions at the POM2 site, we used drip water $\delta^{18}O$ values to calculate a theoretical $\delta^{18}O$ value of the farmed calcite, using the equation given by Tremaine et al. (2011):

$$1000 \ln \alpha = 16.1\,(\pm 0.65) \times 10^3 T^{-1} - 24.6\,(\pm 2.2).$$

The resulting value of -8.5 ± 0.1 ‰ is slightly below the average of -7.9 ‰ measured on calcite farmed at POM2 and POM X sites. Although the 0.6 ‰ offset from generally predicted values could indicate some kinetic fractionation, calcite precipitation can still be considered to have taken place close to equilibrium during the monitored period. However, calculations using the equations in Kim and O'Neill (1997) and Day and Henderson (2011) returned theoretical $\delta^{18}O$ values of -9.2 and -9.6 ‰, respectively, well

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below the measured values on farmed calcite. In this study we base our calculations on the empirical equation of Tremaine et al. (2011), which appear to better characterize in-situ cave conditions.

3.3 Speleothem stable isotope data

The isotopic profiles in Fig. 7 display two common features: an apparent lack of trend during the Early Holocene (8.2–6 ka) and a trend towards higher values during the Middle Holocene (6–4 ka). The δ^{18}O profile returns to relatively stable values during the Late Holocene (4–0 ka), interrupted by a short period (3.2 to 3.0 ka) characterized by low isotope values (Fig. 7).

During the Middle Holocene, the carbon isotope profile shows an ascending trend until 4 ka. The Late Holocene general trend apparently reverses direction towards lighter values (from −9.5 to −10.4 ‰), recording several large fluctuations (Fig. 7).

3.3.1 The δ^{18}O record

The trend observed in δ^{18}O values during the Middle Holocene shows some similarity to the stacked Eastern Mediterranean lacustrine δ^{18}O record (Roberts et al., 2011). To place the Ascunsă Cave stable isotope record in a regional context alongside other speleothem data, we compare our record with δ^{18}O profiles of stalagmites from Poleva Cave (Constantin et al., 2007), Urşilor Cave (Onac et al., 2002) and V11 Cave (Tămaş et al., 2005). We also add δ^{18}O values calculated by McDermott et al. (2011) as representative for low altitude European caves at 22° E longitude (Fig. 8). The data compiled by McDermott et al. (2011) represent the output of a modelled Rayleigh distillation of Atlantic air masses during westerly flow across the European continent. The discrepancy between measured and calculated values suggests that, apart from the dominant Atlantic moisture source, other factors might have played a role in driving the isotopic variability at Ascunsă Cave (e.g. evaporation, multiple vapour sources).

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Poleva Cave shows slightly higher calcite δ^{18}O values after 4 ka (−7.6 ‰) in comparison to > 6 ka (−8.3 ‰) and Constantin et al. (2007) interpret this increase as a general warming trend. At Urşilor Cave (NW Romania), Late Holocene δ^{18}O values are slightly higher (by 0.2 ‰) than during the Middle Holocene and Onac et al. (2002) suggests that an apparent lack of variability at this site reflects relatively stable climate conditions.

Rapid climate change events (Mayewski et al., 2004) such as the 8.2 ka event are not clearly expressed in Romanian speleothem δ^{18}O records (Onac et al., 2002; Tămaş et al., 2005; Constantin et al., 2007). On the contrary, a common negative excursion occurring at ∼ 3.2 ka is recorded in the δ^{18}O time series of Ascunsă and Poleva caves in the Southern Carpathians. This century-long cold event has also been identified in marine records from the Eastern Mediterranean (e.g. Rohling et al., 2002).

3.3.2 Constraining regional temperature change in the speleothem δ^{18}O record with independent temperature reconstructions

Stable oxygen isotopes in speleothems are potentially influenced by local effects, such as cave hydrology and cave ventilation, which may obscure the regional climate signal (Tremaine et al., 2011; Riechelmann et al., 2013). Here, we employ coeval data recorded in more than one cave to account for such potential biases. We specifically address: (1) the general Mid-Holocene trend by comparing the isotopic difference between 2000-yr averaged time intervals between the Early and Late Holocene, from 8 to 6 ka and 4 to 2 ka, respectively (Fig. 8); (2) the absence of an unambiguous 8.2 ka event in isotopic speleothem records from Romania, and (3) the nature of a clear isotope excursion ∼ 3.2 ka in two Southern Carpathian speleothems.

The principal controls of oxygen isotope fractionation during speleothem-calcite precipitation are temperature in the cave and isotopic composition of drip water. Both, directly respond to changes of annual average air temperature above the cave (e.g. Day and Henderson, 2011; Tremaine et al., 2011). In addition, drip water δ^{18}O may also

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record variations in hydrologic climate characteristics, such as rainfall seasonality, evaporation, and concurrent input from different moisture sources (McDermott, 2004; Fairchild et al., 2006; Lachniet, 2009). Assuming that (1) calcite precipitation temperature in the cave reflects the annual average surface air temperature that oscillates very little year around (see also Table 2) and (2) the coldest and warmest months define the range of temperature-controlled oxygen isotope fractionation during condensation of rain, we calculate an expected range for relative changes of δ^{18}O in speleothem records based entirely on temperature variation. For that purpose, we employ pollen-based reconstructions of the annual average surface air temperature (TANN – Temperature ANNual), surface air temperature of the coldest month (MTCO – Mean Temperature of the COldest month), and surface air temperature of the warmest month (MTWA – Mean Temperature of the Warmest month) for two zonal sectors from central Europe and the Mediterranean, respectively (Davis et al., 2003). We use the empirical equation of Tremaine et al. (2011) for temperature-dependent oxygen isotope fractionation during calcite precipitation:

$$1000 \ln \alpha = 16.1 \left(10^3 T^{-1}\right) - 24.6. \tag{1}$$

For the δ^{18}O-temperature relationship in rainwater we use the empirical global mid-latitude relationship suggested by Rozanski et al. (1993):

$$\delta^{18}\mathrm{O}/\Delta T = 0.58\,‰\,°\mathrm{C}^{-1}. \tag{2}$$

For calcite precipitation, $\Delta\delta^{18}\mathrm{O}/\Delta T$ is $\sim -0.18\,‰\,°\mathrm{C}^{-1}$ (Tremaine et al., 2011), therefore the combined temperature effect in speleothem δ^{18}O is dominated by rainfall temperature and resulting changes in drip water δ^{18}O.

We consider rather relative than absolute the changes across the Holocene time intervals of interest. We then compare the calculated temperature-constrained range of relative δ^{18}O variation with the one measured in several Carpathian speleothem records in order to identify the likelihood of additional changes in other climate parameters (e.g. rainfall seasonality, evaporation from the soil, or variable moisture sources and pathways).

If only temperature variability contributed to the observed difference between two time intervals of interest, this change in $\delta^{18}O_c$ will plot inside the range calculated from the pollen input data TANN, MTCO and MTWA. Ideally, the position inside that calculated range should reflect the unchanged annual distribution of rainfall above the cave, but see discussion of uncertainties in the appendix. If any hydrologic factor had changed along with temperature, the observed change of $\delta^{18}O_c$ may fall outside the calculated temperature constrained range. For example, a significant change in rainfall seasonality should result in the observed change of $\delta^{18}O_c$ plotting above the range if the proportion of summer rain increased. The proportional increase of summer rain may of course be the result of additional summer rain, or a reduction of winter rain. Other hydrologic factors, such as a different proportion of moisture from the Mediterranean and the Atlantic, respectively (see Fig. 6), or a different isotopic composition of the sea surface in either two source regions, may also cause the observed $\delta^{18}O_c$ to fall outside the temperature constrained range.

3.3.3 Temperature and hydrology-related changes in speleothem $\delta^{18}O$ records from Romania and the Mediterranean basin

We analyse the broad $\delta^{18}O$ change across the time interval 6–4 ka as outlined above for the Romanian stalagmites and also for a selection of southern European records. For simplicity, this isotopic transition is defined as the difference between the average $\delta^{18}O$ during the 4–2 and 8–6 ka intervals ($\Delta\delta^{18}O_{6-4\,\mathrm{ka}}$).

The pollen-based temperature reconstructions by Davis et al. (2003) divide Europe into six main regions: north-western (NW), north-eastern (NE), central-western (CW), central-eastern (CE), south-western (SW) and south-eastern (SE). The boundary between central and southern zones is 45° N, the boundary between western and eastern zones is 15° E. This places the Alps and much of northern Italy inside the CW zone

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and divides Romania between the CE and SE zone along the Southern Carpathians. Across the last 8000 yr the CW shows a slight winter warming, the CE zone shows only little change, the SW zone shows a 2 °C warming trend for both seasons, and the SE zone shows a 1 °C warming during summer (Davis et al., 2003).

The specific regionally averaged pollen data sets used to calculate isotopic variability in different cave records are summarized in Table 4 and calculation details are given in Table 5. Ambiguities and potential shortcomings of the chosen pollen zones as well as using the Rozanski et al. (1993) empiric relationship between rainfall temperature and its oxygen isotope composition are also discussed in the appendix.

For Ascunsă Cave, which is inside the SE pollen zone of Davis et al. (2003), speleothem $\Delta\delta^{18}O_{6-4\,ka}$ is 0.72 ‰, whereas values expected from the pollen-based temperature reconstruction are between 0.16 ‰ (summer) and −0.05 ‰ (winter) (Fig. 9). This implies that, across the Middle Holocene transition, speleothem $\delta^{18}O$ values at the cave site became higher than expected if controlled by temperature change alone. As $\delta^{18}O$ increases in the majority of observed speleothems from the Eastern Mediterranean domain across the 6–4 ka interval beyond the temperature-controlled amount (Fig. 9), there must have been a common hydrologic change. This may include any combination of change in rainfall seasonality (Lachniet, 2009), local evaporation, a change in the proportion of Atlantic vs. Mediterranean moisture source (Rozanski et al., 1993), or a change in the isotopic composition of the two vapour sources.

To rule out local climate effects, we compare our speleothem record with other isotope records from Poleva (Constantin et al., 2006) and Urşilor (Onac et al., 2002) caves. Considering that the Davis et al. (2003) CE pollen zone is not well constrained near the 45° N latitude in Romania, for Urşilor Cave we use the temperature reconstructions derived from the pollen record of Steregoiu (Feurdean et al., 2008). Figure 8 shows that measured $\Delta\delta^{18}O_{6-4\,ka}$ at Poleva is similar to that at Ascunsă and falls well outside the pollen-temperature constrained range of change, whereas Urşilor falls within the constrained range close to the summer temperature value. Altogether, this suggests that the 6–4 ka transition at Ascunsă marks a significant hydrologic change in southern Romania. The dominantly temperature-related change at Urşilor compared to the southern Romanian cave sites seems to indicate a different Holocene N–S climate evolution across the Southern Carpathians, similar to developments in other parts of Europe at these latitudes (see review in Magny et al., 2013).

To verify the suspected influence of Mediterranean and Atlantic influence on regions of Romania, we calculated similar pollen temperature constrained δ^{18}O values for several cave records close to the Mediterranean, across the same 6–4 ka transition. These records are: Grotte de Clamouse, France (McDermott et al., 1999), in combination with the Davis et al. (2003) SW European temperature time series; Buca della Renella, Italy (Drysdale et al., 2006) and Spannagel Cave, Austria (Vollweiler et al., 2006), each in combination with the Davis et al. (2003) CW European temperature time series; and finally Sofular Cave, Turkey (Fleitmann et al., 2009), Soreq Cave, Israel (Bar-Matthews et al., 2003) and Jeita Cave, Lebanon (Verheyden et al., 2008), each in combination with the Davis et al. (2003) SE European temperature time series. The pollen reconstructions show rising temperatures throughout the year for the SW zone (Davis et al., 2003), whereas in the CW zone, increasing winter temperatures offset decreasing summer temperatures. The results are shown in Fig. 9. It is apparent that isotope values from the Western Mediterranean (Clamouse) and CW Europe south of the Alpine divide (Renella and Spannagel) show a change in δ^{18}O that is explained almost entirely by pollen-reconstructed temperature changes. Only at the Renella site, a small hydrologic influence could be argued for, as the observed change is close to the summer end of the range (Fig. 9), whereas present day rainfall is dominated by the winter season (Scholz et al., 2012). On the other hand, sites in Turkey (Sofular), Israel (Soreq), Lebanon (Jeita), and southern Romania (Poleva and Ascunsă) that are influenced by the Eastern Mediterranean plot well above the temperature-explained change. Thus, it appears that across the Middle Holocene transition, the entire Eastern Mediterranean realm underwent a significant moisture-balance change in combination with some temperature forcing, whereas in the Western Mediterranean such hydrologic change is not apparent.

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The large scale pattern across the Mediterranean suggests different forcing of climate change over the West and the East respectively, but also a common cause of hydrologic change over the Eastern Mediterranean domain. In principle, three aspects may contribute to the observed hydrologic change in the East: (1) different rainfall seasonality, (2) a change in proportion of moisture source, and (3) a different isotopic composition of the moisture sources. Higher δ^{18}O values could be the result of more rain falling in the warmer months during Late Holocene, but this is unlikely because of large scale subsidence over the Eastern Mediterranean region due to the Asian monsoon (Rodwell and Hoskins, 1996; Staubwasser et al., 2006). As a result, there are virtually no rainfalls during summer in the Levant, and are significantly reduced over SE Europe. The summer months are those with lowest rainfall in SW Romania. Subsidence and accompanying low humidity over the Eastern Mediterranean may, however, have increased evaporation, which would be in agreement with rising summer temperatures in SE Europe across the Holocene (Davis et al., 2003). Evaporation in soil and epikarst drives drip water δ^{18}O towards more positive values, resulting in higher δ^{18}O in speleothem calcite (Bar-Matthews et al., 1996; Fairchild et al., 2006).

Alternatively, the proportion of summer rain could have been also higher if winter rainfall decreased. McDermott et al. (2011) suggested lower rainout efficiency during winter along a West-East transect across central Europe. However, higher winter temperatures only in the Western Mediterranean region (Davis et al., 2003) would increase the temperature gradient between SW Romania and the source of cyclones in the Gulf of Genoa, possibly leading to increased rainout efficiency in South Europe.

Another factor that may have added to the observed increase in speleothem δ^{18}O is a change in the isotopic composition of the Mediterranean mixed layer. A steadily increasing δ^{18}O by almost 1 ‰ is recorded in tests of surface dwelling foraminifera from the Aegean Sea between 8 and 4 ka BP, which is uncorrelated with the abundance of cold water species (Rohling et al., 2002). In the Adriatic Sea, an increase by 0.5 ‰ was recorded at the same time (Siani et al., 2010). As such, a combination of warmer summer temperatures, enhanced evaporation from the soil, and higher δ^{18}O of the

Eastern Mediterranean moisture source are currently the favoured explanation for the observed increase in $\delta^{18}O$ of speleothems from the Eastern Mediterranean domain.

3.3.4 The $\delta^{13}C$ record

Interpretation of speleothem $\delta^{13}C$ data is generally hampered by a host of local factors such as changes in soil CO_2 production and content, closed versus open system dissolution of carbonates in the soil/epikarst system, residence time and mixing of waters along the pathway to the drip point, or solution degassing (Hendy, 1971; Bar-Matthews et al., 1996; Fairchild et al., 2006).

Percolating water degassing could be greater during certain periods at POM2 sampling site, as the CO_2 content of the cave's atmosphere drops from $\sim$1800 ppm in November–December to $\sim$1000 ppm in April–May. This seasonal variation in the CO_2 content of cave air is likely the combined result of soil CO_2 productivity and cave ventilation (Spötl et al., 2005; Kowalczk and Froelich, 2010; Frisia et al., 2011; Tremaine et al., 2011; Riechelmann et al., 2013).

At the POM A site, which is the shallowest and closest to the entrance, the pCO_2 reaches a minimum value of 760 ppm, well above values of outside air (between 200 and 310 ppm). The two deeper sites, POM2 and POM B, show even less ventilation, with minimal values of 960 ppm. This indicates that cave ventilation is moderate (although continuous) at Ascunsă Cave.

Supposing that the cave ventilation regime remained unchanged during the Middle Holocene, cave air pCO_2 was probably controlled mostly by soil/vegetation dynamics. If so, higher speleothem $\delta^{13}C$ values are indicative of reduced CO_2 input from the soil and/or prior precipitated calcite (Fairchild et al., 2000). These two processes could be the result of increasing drought conditions and might have been responsible for producing the upward trend observed in $\delta^{13}C$ values during the Middle Holocene. This situation is consistent with the increasing drought implied by oxygen isotopes in our stalagmite.

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3.3.5 The 8.2 and 3.2 ka events

The 8.2 ka climate change event (Alley et al., 1997; Rohling and Pälike, 2005) is one of the most prominent events of environmental change in the Holocene. Pollen assemblages from northern Romania (Feurdean et al., 2008), Macedonia (Bordon et al., 2009) and Greece (Pross et al., 2009), testate amoebae from northern Romania (Schnitchen et al., 2006), speleothem carbon stable isotopes from Israel (Bar-Matthews et al., 2000) and marine faunal composition from the Aegean Sea (Rohling et al., 2002) document a decrease in winter temperature and precipitation, while summer conditions remained rather stable. Similar to other Romanian stalagmites (Tămaş et al., 2005; Constantin et al., 2007), the POM2 δ^{18}O and δ^{13}C records do not show significant variations across the 8.2 ka event. The only indication of changing environmental conditions is that the growth rate was 8 times higher during this event compared to the rest of the Holocene in the Ascunsă Cave.

Figure 10 shows a comparison of $\Delta\delta^{18}$O for pollen temperature-constrained and measured oxygen isotope values for the 8.2 and 3.2 ka events. For the 8.2 ka event, $\Delta\delta^{18}$O is calculated as the difference between a 500-yr interval succeeding the event (8.1–7.6 ka) and the event itself (8.3–8.1 ka). Here we used pollen-based temperature reconstructions as follows: for V11 Cave, those from the Steregoiu peat bog in northern Romania (Feurdean et al., 2008), and for Ascunsă Cave, the Lake Maliq in Macedonia (Bordon et al., 2009).

The shift in isotopic values after the 8.2 ka event at Ascunsă Cave is seemingly explained by the pollen-based temperature rise (Feurdean et al., 2008; Bordon et al., 2009). Nevertheless, as calcite values from Ascunsă are closer to summer values, a hydrological influence on these values could be argued for. As the annual reconstructed pollen temperature rose after the 8.2 ka event, so did the cave temperature. Supposing that rainfall $\Delta\delta^{18}\text{O}/\Delta T$ slope was identical to present day, the decreasing δ^{18}O values change resulting from a warmer cave atmosphere must have been offset by the increasing of the δ^{18}O values stemming from higher rainfall temperature in

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winter. Yet, no significant change in $\delta^{18}O$ is observed in the record. Observed $\delta^{18}O$ values fall within but close to the low $\delta^{18}O$ summer end of the calculated temperature-constrained range (Feurdean et al., 2008; Bourdon et al., 2009). As such, the lack of a clear $\delta^{18}O$ signal across the 8.2 ka event can also be explained by a relative increase of the amount of winter infiltration after the event, thereby increasing the proportion of lower $\delta^{18}O$ in drip water. A possible scenario for the 8.2 ka cold event itself would be that overall lower drip water $\delta^{18}O$ values associated with lower annual temperatures were offset by a larger proportion of summer rainfall.

The 0.6 ‰ increase in $\delta^{18}O$ values after the 3.2 ka event in the Ascunsă record was analysed by comparing the periods 3.0–2.5 ka with 3.2–3.0 ka. These intervals are similar at Poleva (Constantin et al., 2007), showing a comparable structure, although with a small difference in chronology. At Poleva, the two intervals are 2.95–2.53 and 3.07–3.01. The difference in timing is most probably a result of differences in age determinations, while still within uncertainties. The magnitude of rising isotope values after the event is outside the pollen-defined change in $\delta^{18}O_c$ due to temperature, thus it likely reflects a hydrologic change. The interval is coeval with events documented in both archaeological and palaeoclimate records around the Eastern Mediterranean (see review in Drake, 2012) that may have led to the demise of the Late Bronze Age (Kaniewski et al., 2010). Associated to this cold event, a decrease of the Aegean Sea winter surface temperatures was documented by Rohling et al. (2002), whereas a drop of the Dead Sea level reflects drier conditions in the Eastern Mediterranean (Migowski et al., 2006). In northern Romania, testate amoebae data indicate a dry phase between 3.39 and 3.03 ka (Schnitchen et al., 2006), while pollen-based temperature reconstructions clearly show decreased annual and winter values (Feurdean et al., 2008).

The temperature-constrained isotopic change calculated using pollen reconstructed temperatures from Maliq Lake (Bordon et al., 2009) lies between −0.2 ‰ for the cold season and 0.4 ‰ for summer months. The measured value from Ascunsă Cave is 0.6 ‰; this offset towards higher values possibly indicates that hydrological processes were responsible for this enrichment. The Mediterranean plays an important role as

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winter moisture source in south-western Romania (Bojariu and Paliu, 2001). The ^{18}O depleted waters associated with the 3.2 ka event could be the combined result of lower temperatures at the cave site and a decreased Mediterranean input resulting from less winter evaporation of sea surface waters. Reduced sea surface temperatures also imply more depleted $\delta^{18}O$ values for winter moisture contributing to the downtrend observed in the Ascunsă isotope time series. After the 3.2 ka event, as cave temperature rose, the isotopic processes affecting calcite precipitation would have lowered $\delta^{18}O$ values. However, this effect might have been overwritten by raising summer temperatures, possibly accompanied by evaporation, similar to developments observed at the Middle Holocene transition.

4 Conclusions

The stable isotope record of Ascunsă Cave in southern Romania was used to identify centennial to millennial climate change during the Holocene in this area. Between 6 and 4 ka, $\delta^{18}O$ gradually shifted towards higher values and we show that the Atlantic source effect was not the main isotopic driver for this well-defined isotopic shift.

By using a new approach to discriminate between the effects of temperature and hydrology on speleothem $\delta^{18}O$ values, we demonstrate that this shift not only reflects rising regional temperatures as documented by pollen assemblages (Davis et al., 2003), but also a combination of hydrologic influences.

The approach presented in this study relies on using pollen-based temperature reconstructions to constrain temperature-driven isotopic changes of speleothem calcite. This method can use any independent temperature reconstructions and considers the isotopic fractionation occurring during water vapour condensation and calcite precipitation. A constrained range of temperature-driven isotopic changes between winter and summer is obtained, and departures from this range suggest that additional factors ultimately control the isotopic variability over the studied period.

Using this approach we find that the Middle Holocene enrichment in SW Romania was 0.6 ‰ greater than the maximum values, likely associated with rising temperatures. We further extended the calculation to other speleothem records in the Eastern and Western Mediterranean and western Romania (Urşilor Cave). In the Atlantic-dominated western Romania, isotopic change largely followed temperature variations, revealing different climate response of these two regions separated by the Southern Carpathian mountain range.

We also show that during the Middle Holocene, δ^{18}O in Western Mediterranean speleothems responded mostly to temperature change. At the same time, other processes such as enhanced evaporation rates or gradual enrichment in isotopic composition of surface waters could have contributed to the observed isotope change in areas influenced by the Eastern Mediterranean climate.

We analysed two rapid climate changes, at 8.2 and 3.2 ka. At Ascunsă Cave, the 8.2 ka event is characterized by a growth rate 5–6 times greater than during the rest of the Holocene. Nevertheless, the δ^{18}O and δ^{13}C values show low variability, whereas the 3.2–3.0 ka period is well defined by a 1.5 ‰ depletion in δ^{18}O. The low isotopic variability during the 8.2 ka event seems to reflect only temperature variations, but hydrologic conditions such as relatively more summer vs. winter rainfall at Ascunsă and V11 caves cannot be ruled out.

During the 3.2 ka event, the thermic and hydrological impact on speleothem isotope values is obvious at Ascunsă Cave, reflecting decreased winter temperature and a lower and perhaps isotopically-lighter Mediterranean moisture input.

Appendix A

Calculation method of temperature constrained isotope values

The calculation of the temperature-related part of an observed change in speleothem calcite δ^{18}O from pollen-based temperature reconstructions relies on two basic

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assumptions: (1) the cave temperature reflects the annual average surface air temperature and only fluctuates very little around that value (i.e. temperatures in the Ascunsă Cave chamber from which the POM2 stalagmite was collected vary by 0.6 °C over the year); (2) the coldest and warmest month reasonably define the range of temperature-controlled oxygen isotope fractionation during rainfall. Currently, this cannot be tested for the Ascunsă Cave site due to a lack of a continuous recording of stable isotopes in precipitation. However, this assumption is based on information from other European station recordings (Rozanski et al., 1993).

In the following, we only consider temperature-related aspects contributing to an observed isotopic change, $\Delta\delta^{18}O$. Generally, $\delta^{18}O$ in calcite is determined by the calcification temperature and the isotopic composition of ambient water (Epstein et al., 1953), the latter reflecting rain formation temperature among other hydrologic factors (Rozanski et al., 1993). Consequently, $\Delta\delta^{18}O$ can be divided into $\Delta\delta^{18}O_c$ – the contribution due to changing calcification temperature – and $\Delta\delta^{18}O_w$ – the contribution due to changing rain temperature. The relative change of $\delta^{18}O$ between two time intervals, t_1 and t_2, in a speleothem is:

$$\Delta\delta^{18}O_{t_1-t_2} = \Delta\delta^{18}O_{c(t_1-t_2)} + \Delta\delta^{18}O_{w(t_1-t_2)} = \delta^{18}O_{t_1} - \delta^{18}O_{t_2}. \tag{A1}$$

Likewise, the change in pollen temperature anomaly (Davis et al., 2003) is:

$$\Delta TA^{pollen}_{t_1-t_2} = TA^{pollen}_{t_1} - TA^{pollen}_{t_2}. \tag{A2}$$

The absolute pollen-derived temperature at any given time, t, is:

$$T^{pollen}_t = T_{today} + TA^{pollen}_t. \tag{A3}$$

The empirical fractionation factor α for oxygen isotopes between water and calcite is defined as (Tremaine et al., 2011):

$$1000 \ln \alpha = 16.1\left(10^3 T^{-1}\right) - 24.6. \tag{A4}$$

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The fractionation factor is related to measured values for $\delta^{18}O_c$ and $\delta^{18}O_w$ by (e.g. Sharp, 2007):

$$1000 \ln \alpha \approx \delta^{18}O_c - \delta^{18}O_w \quad (A5)$$

where we express both $\delta^{18}O$ values in relation to SMOW. In the case of $\delta^{18}O_w = 0$, Eq. (A5) reduces to:

$$1000 \ln \alpha \approx \delta^{18}O_c. \quad (A6)$$

Although this approximation would deviate somewhat from the true relationship at the given 25–30 ‰ difference between the two $\delta^{18}O$ values, most of the error cancels out when calculating:

$$\Delta\delta^{18}O_{c(t_1-t_2)} = \delta^{18}O_{c(t_1)} - \delta^{18}O_{c(t_2)}. \quad (A7)$$

From Eqs. (A3), (A4), (A6) and (A7) we have:

$$\Delta\delta^{18}O_{c(t_1-t_2)} = 16.1 \left(\frac{1}{T_{t_1}^{\text{pollen}}} - \frac{1}{T_{t_2}^{\text{pollen}}} \right) 1000. \quad (A8)$$

To constrain the range of temperature-related variability of $\delta^{18}O_w$ we use the empirical relationship of 0.58 ‰ ($\delta^{18}O$)/°C for mid-latitudes (Rozanski et al., 1993):

$$\Delta\delta^{18}O_{w(t_1-t_2)} = 0.58\,\Delta TA_{t_1-t_2}^{\text{p-seasonal}} \quad (A9)$$

with summer (MTWA) and winter (MTCO) temperatures from the Davis et al. (2003) pollen-based reconstructions for $\Delta TA^{\text{p-seasonal}}$. Inserting Eqs. (A8) and (A9) into Eq. (A1) yields the pollen-constrained range of change for $\delta^{18}O$ in speleothem calcite – after conversion to the PDB scale – that may be explained by temperature variability between two defined time intervals (Fig. 9).

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The uncertainty of the pollen-constrained temperature range can in principle be estimated by a full propagation of errors, but these are not generally available for the pollen reconstruction by Davis et al. (2003). Accuracy will obviously strongly depend on the choice of pollen-based comparison datasets. Davis et al. (2003) recommend that caution should be taken when comparing data from individual sites with regional reconstructions within which large local differences may occur. Uncertainty of pollen temperatures will result in shifting, shrinking and expanding of the δ^{18}O range constrained by those temperatures. The authors also outline the fact that the regions of Europe were chosen arbitrarily and the continuum of the climate change might not be well expressed between them.

Results from Eq. (A9) will be affected by spatial and temporal variability of the 0.58 ‰ (δ^{18}O)/°C slope defined by Rozanski et al. (1993). However, the observation that much of the change in δ^{18}O across the 4–6 ka transition in the Eastern Mediterranean (Fig. 9) requires a hydrologic component is reasonably robust. The distance between the temperature-constrained interval and the observed δ^{18}O value in Fig. 9 would approximately shrink by half if the 0.58 slope value would be twice as high, which is not observed in any of the individual stations summarized by Rozanski et al. (1993).

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Table 1. Results of the U-Th measurements of POM2 samples.

Sample ID	Distance from bottom (cm)	^{238}U ($ng\,g^{-1}$)	^{232}Th ($ng\,g^{-1}$)	^{230}Th ($ng\,g^{-1}$)	$[^{230}Th/^{232}Th]$ activity ratio	$(^{232}Th/^{238}U)$ activity ratio	$(^{230}Th/^{238}U)$ activity ratio	$(^{234}U/^{238}U)$ activity ratio	Uncorrected age (ka)	Corrected age (ka)	Corrected $(^{234}U/^{238}U)_{initial}$ activity ratio
POM 09-2/top	76.35	18.26 ± 0.11	12.155 ± 0.074	3.951E-05 ± 3.44E-06	0.61 ± 0.05	2.178E-01 ± 6.002E-04	1.322E-01 ± 1.051E-02	1.791E+00 ± 7.699E-03	8.330 ± 0.685	0.098 ± 4.354	1.9098 ± 0.070
POM 09-2/I	63.35	18.94 ± 0.09	0.330 ± 0.003	1.122E-05 ± 3.29E-07	6.34 ± 0.18	5.706E-03 ± 4.247E-05	3.620E-02 ± 1.021E-03	2.087E+00 ± 6.154E-03	1.908 ± 0.055	1.729 ± 0.104	2.0960 ± 0.006
POM 09-2/III	62.85	18.99 ± 0.10	8.833 ± 0.044	2.264E-05 ± 6.54E-07	0.48 ± 0.01	1.522E-01 ± 4.452E-04	7.281E-02 ± 1.867E-03	1.848E+00 ± 6.245E-03	4.377 ± 0.115	–	–
POM 09-2/VI	53.75	19.11 ± 0.10	10.289 ± 0.054	2.100E-05 ± 6.81E-07	0.38 ± 0.01	1.761E-01 ± 4.941E-04	6.713E-02 ± 2.375E-03	1.969E+00 ± 7.584E-03	3.779 ± 0.136	–	–
POM 09-2/II	53.25	21.07 ± 0.11	0.605 ± 0.005	1.945E-05 ± 4.97E-07	6.00 ± 0.15	9.393E-03 ± 7.452E-05	5.639E-02 ± 1.441E-03	2.041E+00 ± 9.131E-03	3.052 ± 0.080	2.751 ± 0.167	2.0555 ± 0.010
POM 09-2/V	42.65	19.12 ± 0.09	0.588 ± 0.005	2.595E-05 ± 4.27E-07	8.24 ± 0.14	1.007E-02 ± 8.008E-05	8.293E-02 ± 1.451E-03	2.091E+00 ± 6.276E-03	4.405 ± 0.080	4.091 ± 0.171	2.1102 ± 0.007
POM 09-2/IV	42.25	19.62 ± 0.08	0.705 ± 0.006	2.775E-05 ± 5.81E-07	7.35 ± 0.15	1.175E-02 ± 9.093E-05	8.640E-02 ± 1.596E-03	2.066E+00 ± 8.421E-03	4.649 ± 0.090	4.278 ± 0.199	2.0868 ± 0.009
POM 09-2/VIII	32.3	42.96 ± 0.15	0.378 ± 0.004	7.403E-05 ± 9.71E-07	36.55 ± 0.52	2.794E-03 ± 2.624E-05	1.053E-01 ± 1.252E-03	1.849E+00 ± 4.267E-03	6.373 ± 0.079	6.275 ± 0.092	1.8658 ± 0.0044
POM 09-2/base	20.65	29.44 ± 0.17	0.597 ± 0.006	6.075E-05 ± 9.79E-07	19.00 ± 0.31	6.636E-03 ± 6.077E-05	1.261E-01 ± 1.884E-03	1.815E+00 ± 5.368E-03	7.820 ± 0.123	7.583 ± 0.166	1.8361 ± 0.006
POM 09-2/B	16.4	39.99 ± 0.20	0.245 ± 0.003	8.434E-05 ± 1.89E-06	64.38 ± 1.29	2.001E-03 ± 1.936E-05	1.289E-01 ± 2.710E-03	1.798E+00 ± 4.065E-03	8.077 ± 0.176	8.004 ± 0.180	1.8176 ± 0.004
POM 09-2/C	7.5	49.04 ± 0.25	0.228 ± 0.002	1.044E-04 ± 1.12E-06	85.37 ± 0.90	1.524E-03 ± 1.058E-05	1.301E-01 ± 1.302E-03	1.759E+00 ± 3.910E-03	8.346 ± 0.089	8.290 ± 0.092	1.7776 ± 0.004
POM 09-2/A	4.6	24.02 ± 0.11	0.097 ± 0.001	4.984E-05 ± 8.74E-07	95.85 ± 1.71	1.323E-03 ± 1.147E-05	1.268E-01 ± 2.200E-03	1.773E+00 ± 4.037E-03	8.062 ± 0.146	8.013 ± 0.148	1.7911 ± 0.004
POM 09-2/XXII	2.7	28.59 ± 0.09	0.140 ± 0.002	6.261E-05 ± 9.18E-07	83.63 ± 1.37	1.552E-03 ± 1.992E-05	1.338E-01 ± 1.928E-03	1.816E+00 ± 5.327E-03	8.311 ± 0.126	8.255 ± 0.129	1.8361 ± 0.0054
POM 09-2/XXIII	1.45	31.08 ± 0.10	0.190 ± 0.002	7.192E-05 ± 1.06E-06	71.51 ± 1.08	1.918E-03 ± 2.125E-05	1.414E-01 ± 2.172E-03	1.916E+00 ± 5.269E-03	8.321 ± 0.134	8.256 ± 0.138	1.9387 ± 0.0054
POM 09-2/D	0	20.72 ± 0.12	0.406 ± 0.004	1.060E-04 ± 1.17E-06	48.78 ± 0.53	6.407E-03 ± 4.260E-05	3.126E-01 ± 3.102E-03	2.097E+00 ± 4.978E-03	17.384 ± 0.190	17.189 ± 0.206	2.1563 ± 0.006

Table 2. Spot measurements of physical climate parameters in Ascunsă Cave, including water stable isotopes.

Date	Air T (°C)	RH (%)	$p\text{CO}_2$ (ppm)	δ^{18}O (‰, SMOW)	δD (‰, SMOW)
POM A					
13 Jul 2012	7.5	93.7	1300	N/A	N/A
17 Oct 2012	N/A	N/A	N/A	−10.39	−69.63
30 Nov 2012	8.2	92.0	1520	N/A	N/A
4 Jan 2013	7.8	89.4	1050	−10.36	−68.91
28 Feb 2013	7.2	N/A	760	−10.39	−68.87
20 Apr 2013	7.5	96.8	870	−10.83	−72.43
26 May 2013	8.4	92.3	1070	N/A	N/A
21 Sep 2013	8.6	90.25	1060	N/A	N/A
2 Nov 2013	8.7	89.3	1710	N/A	N/A
POM2					
13 Jul 2012	8.2	94.0	1280	N/A	N/A
17 Oct 2012	N/A	N/A	N/A	−10.571	−70.74
30 Nov 2012	8.2	94.2	1740	−10,582	−70.54
4 Jan 2013	8.1	94.1	1770	−10.517	−70.35
28 Feb 2013	7.6	N/A	1400	−10.596	−70.67
20 Apr 2013	7.9	96.8	960	−10.760	−71.72
26 May 2013	8.8	92.2	1150	N/A	N/A
21 Sep 2013	8.3	94.68	1360	N/A	N/A
2 Nov 2013	8.6	91.25	1820	N/A	N/A
POM B					
13 Jul 2012	8.4	93.1	1300	N/A	N/A
17 Oct 2012	N/A	N/A	N/A	−10.68	−71.46
30 Nov 2012	8.5	93.0	1880	−10.39	−70.01
4 Jan 2013	8.6	92.0	1660	−10.46	−69.77
28 Feb 2013	7.8	N/A	1360	−10.37	−69.15
20 Apr 2013	8.6	94.3	1110	−10.94	−73.24
26 May 2013	9.6	88.6	1270	N/A	N/A
21 Sep 2013	8.5	93.95	1550	N/A	N/A
2 Nov 2013	8.6	93.3	2010	N/A	N/A

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Table 3. Stable isotope values of farmed calcite in Ascunsă Cave.

Sample	$\delta^{13}C$	$\delta^{18}O$
POM A Sep 2010–Jan 2011	−10.657	−8.264
POM A Jan 2011–Jul 2012	−9.455	−7.510
POM A Jul 2012–Oct 2012	−9.485	−7.826
POM2 Jan 2011–Jul 2012	−10.403	−7.550
POM2 Jul 2012–Oct 2012	−10.369	−7.844
POM2 Dec 2012– Jan 2013	−10.434	−7.964
POM2 Jan 2013–Apr 2013	−11.129	−8. 097
POM X Sep 2010–Jan 2011	−9.801	−8.312
POM X Jan 2011–Jul 2012	−10.427	−7.877
POM X Jul 2012–Oct 2012	−9.594	−7.780
POM X Oct 2012–Dec 2012	−10.234	−7.912
POM X Dec 2012–Feb 2013	−10.009	−7.869
POM X Feb 2013–Apr 2013	−10.360	−7.915

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Table 4. Regional pollen datasets (after Davis et al., 2003) used for the calculation of the temperature effect on speleothem isotopic values between 6 and 4 ka.

Pollen region	Average T 6–8 ka	Average T 2–4 ka	ΔT 4–6 ka
TANN CW Europe	−0.26	−0.08	0.18
TANN SE Europe	−0.51	0.04	0.56
TANN CE Europe	1.29	1.02	−0.27
TANN SW Europe	−2.03	−0.82	1.21
MTWA CW Europe	0.32	0.18	−0.13
MTWA SE Europe	−0.88	−0.41	0.47
MTWA CE Europe	0.36	0.10	−0.26
MTWA SW Europe	−1.72	−0.67	1.05
MTCO CW Europe	−0.49	0.37	0.87
MTCO SE Europe	0.20	0.31	0.11
MTCO CE Europe	0.29	0.12	−0.16
MTCO SW Europe	−1.51	−0.38	1.13

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Table 5. Calculation results of temperature constrained speleothem isotope values for the mid-Holocene transition.

Site	Average 6–8 ka	Average 2–4 ka	$\Delta^{18}O$ speleothem 4–6 ka	Cave T °C	Annual ΔT 4–6 ka	Calcite $\Delta^{18}O_{4-6\,ka}$	Summer ΔT 4–6 ka	Winter ΔT 4–6 ka	$\delta^{18}O_{drip\ Summer\ 4-6\,ka}$	$\delta^{18}O_{drip\ Winter\ 4-6\,ka}$	$\Delta^{18}O$ Summer VPDB	$\Delta^{18}O$ Winter VPDB
Urşilor	−7.84	−7.48	0.36	10.0	0.6	−0.12	0.91	0.33	0.53	0.19	0.40	0.07
Ascunsă	−8.69	−7.97	0.72	8.0	0.55	−0.11	0.47	0.11	0.27	0.06	0.16	−0.05
Poleva	−8.26	−7.62	0.64	10.0	0.55	−0.11	0.47	0.11	0.27	0.06	0.16	−0.05
Sofular	−8.53	−8.12	0.41	13.3	0.55	−0.11	0.47	0.11	0.27	0.06	0.16	−0.04
Soreq	−5.91	−5.40	0.51	18.0	0.55	−0.10	0.47	0.11	0.27	0.06	0.16	−0.04
Jeita	−5.38	−4.78	0.60	22.0	0.55	−0.10	0.47	0.11	0.27	0.06	0.17	−0.04
Renella	−3.96	−3.94	0.02	12.0	0.55	−0.11	0.47	0.11	0.27	0.06	0.16	−0.04
Clamouse	−4.92	−4.56	0.36	14.5	1.21	−0.24	1.05	1.13	0.61	0.66	0.36	0.41
Spannagel	−7.81	−7.64	0.17	1.9	0.55	−0.12	0.47	0.11	0.27	0.06	0.15	−0.05

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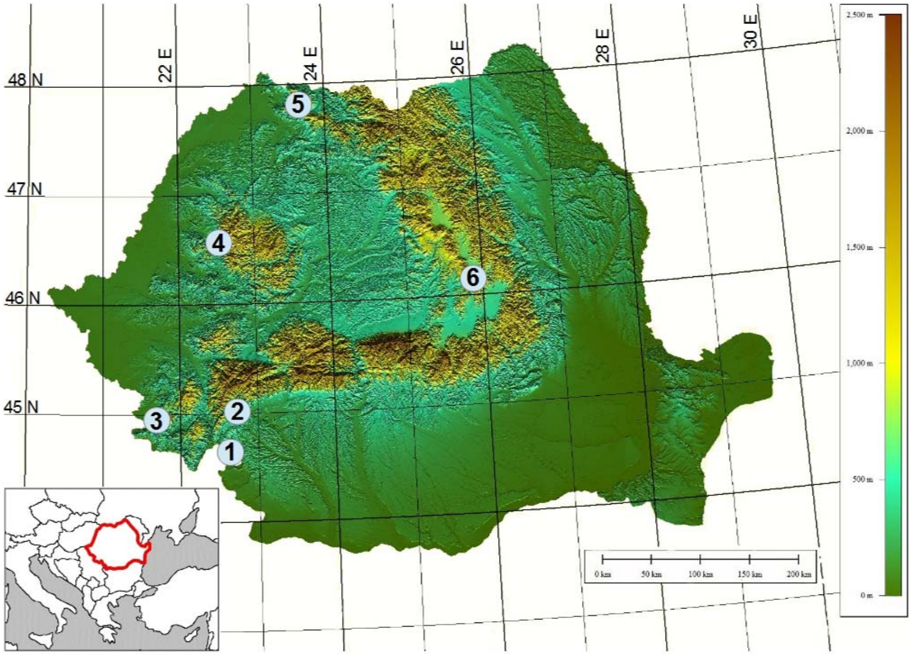

Fig. 1. Location of Romanian palaeoclimate records and meteorological stations mentioned in text: 1 – Drobeta meteorological station; 2 – Ascunsă Cave; 3 – Poleva Cave; 4 – Urşilor Cave, V11 Cave and Stâna de Vale meteorological station; 5 – Steregoiu peat-bog; 6 – Sfânta Ana Lake.

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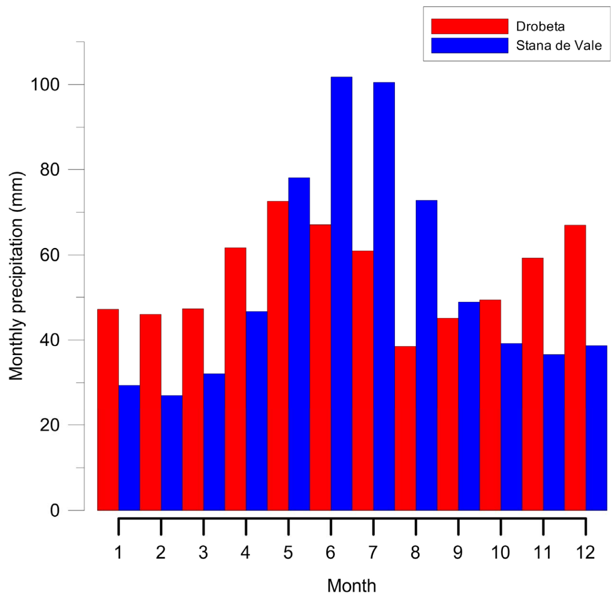

Fig. 2. Average monthly precipitation quantities at stations Drobeta and Stâna de Vale (data from Dragotă and Baciu, 2008).

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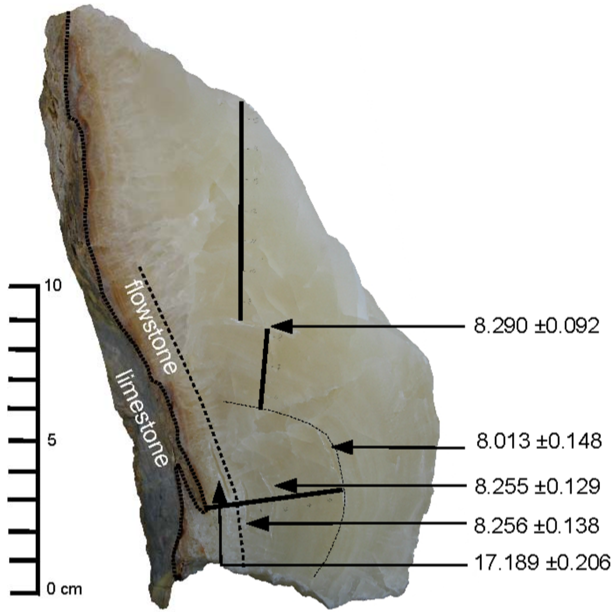

Fig. 3. Base of stalagmite POM2 (ages are given in ka).

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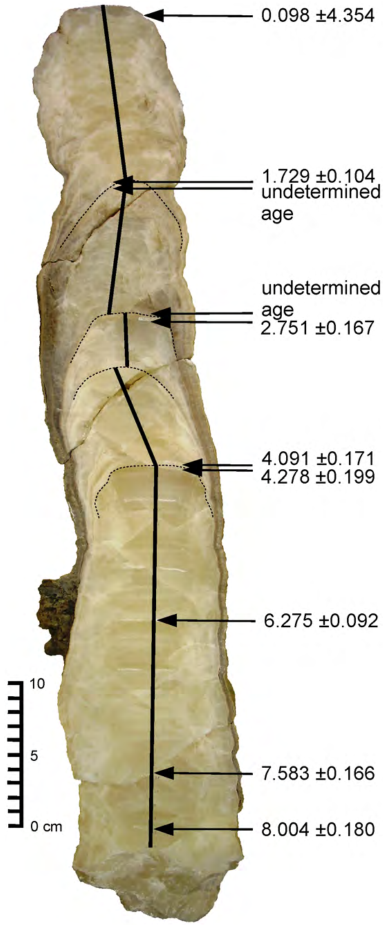

Fig. 4. Upper part of stalagmite POM2 (ages are given in ka).

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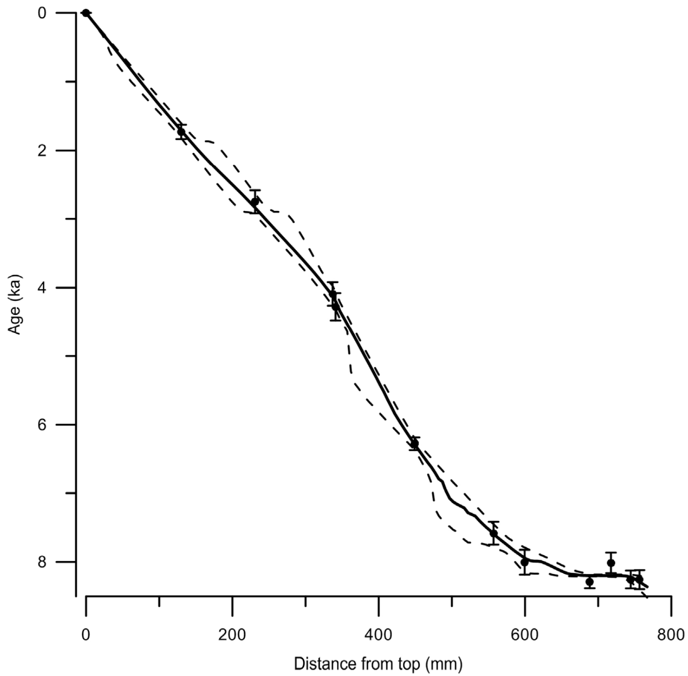

Fig. 5. The growth model (solid line) of stalagmite POM2 with 95 % confidence intervals (dashed lines).

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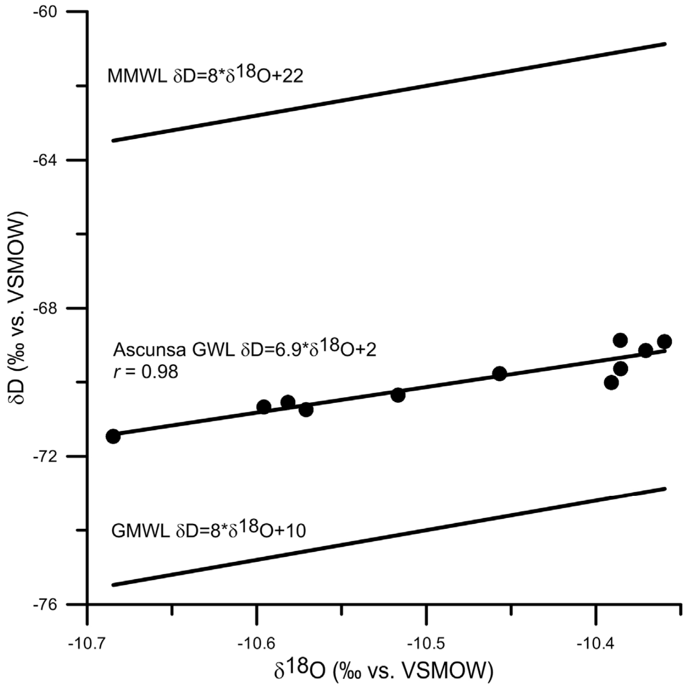

Fig. 6. Comparison between GMWL – global (Craig, 1961) and MMWL – Mediterranean meteoric water lines (Gat and Carmi, 1970), as well as Ascunsă Cave groundwater line.

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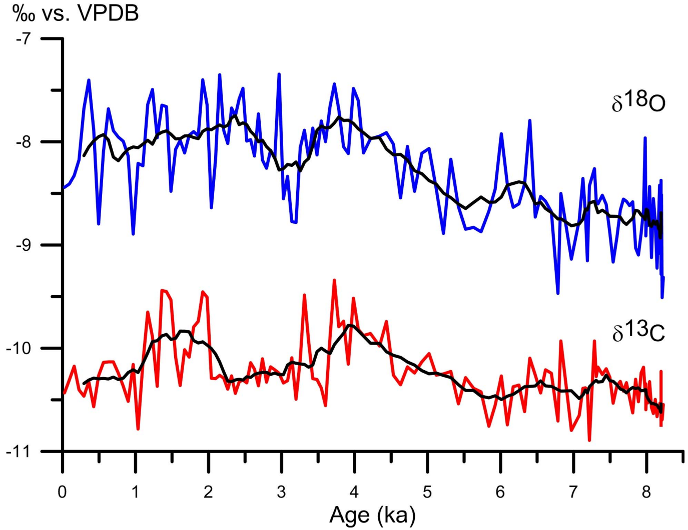

Fig. 7. $\delta^{18}O$ and $\delta^{13}C$ profiles of stalagmite POM2 with 9-point smoothed values (black).

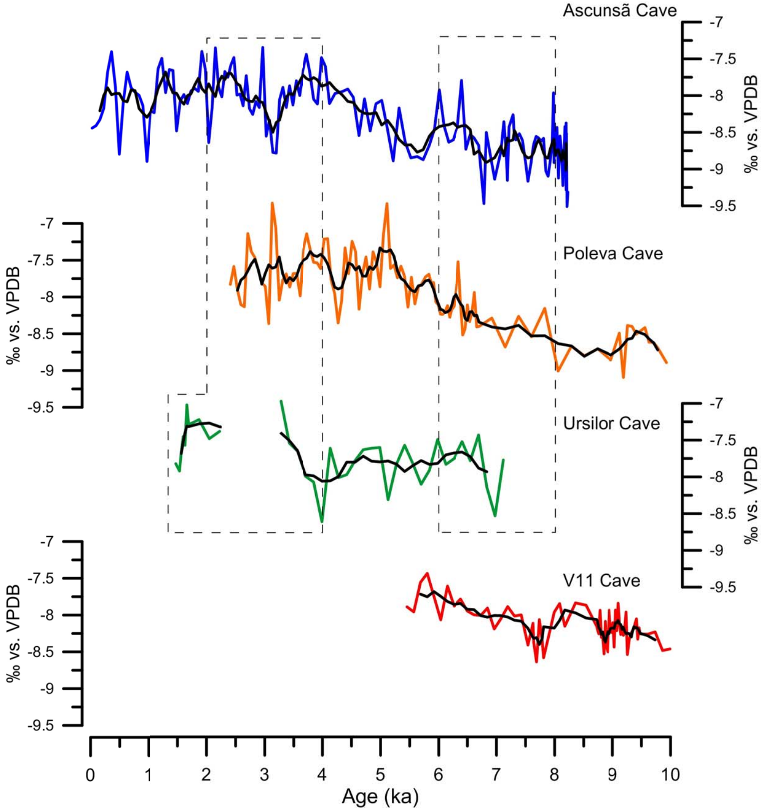

Fig. 8. Comparison between $\delta^{18}O$ records from Ascunsă, Poleva, Urşilor and V11 caves. Isotopic values predicted by McDermott et al. (2011) for low altitude caves at 22° E longitude are represented as dashed line. Dashed line boxes represent the time windows (2–4 and 6–8 ka) for which the average isotopic values were calculated.

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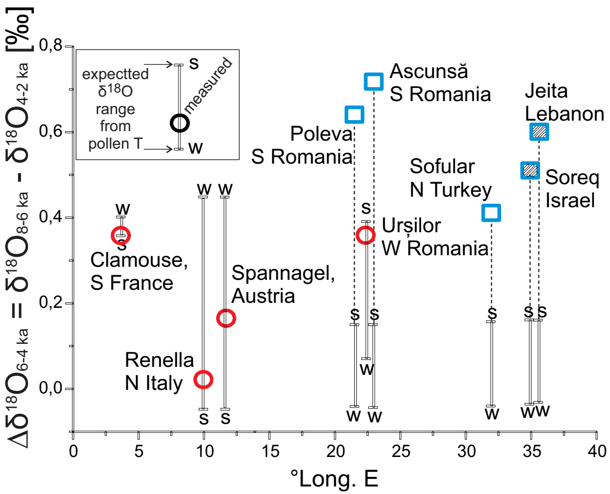

Fig. 9. Comparison of isotopic changes in stalagmites across different longitudes in Europe and predicted isotopic change in winter and summer precipitated calcite in the interval 6–4 ka.

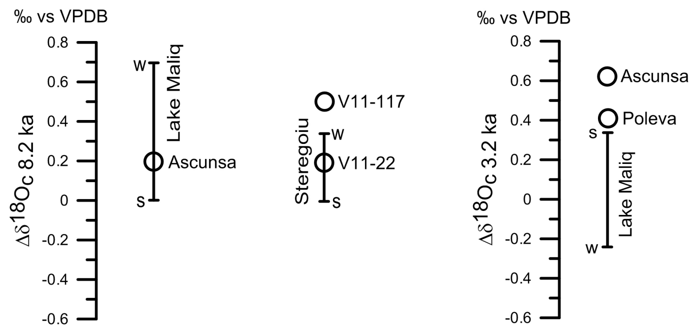

Fig. 10. Comparison of isotopic changes in stalagmites from Ascunsă, Poleva and V11 caves with expected oxygen isotope change constrained by pollen temperature reconstruction in winter and summer, for the 8.2 ka event (left) and the 3.2 ka event (right). $\Delta\delta^{18}O$ as defined in the text.

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